

Preparation Containing Gypsum from Flue Gas Desulphurization, Methodfor Using Same and Use Thereof

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FIELD OF THE INVENTION

This invention relates to a preparation containing a water-soluble polymer or a water-dispersible polymer or a mixture of two or more thereof and gypsum particles from waste-gas desulfurization plants (WDP gypsum) with a value for the particle size distribution x50 of 13 to 500  $\mu\text{m}$ , to a process for its production and to its use.

BACKGROUND OF THE INVENTION

Inorganic inert fillers and polymeric materials are often used in admixture with one another. Depending on the ratio by weight of filler to polymeric material, properties can be attributed to the end product obtainable from such a mixture that would be very difficult or even impossible to achieve using a material which consisted of only one of the components of the mixture (i.e. only polymer or only filler). The combination of inorganic inert filler and polymers is often aimed at and used not least because both materials have very different property profiles of which the combination is not only desirable but actually necessary for many applications.

Fillers which generally consist of a number of individual loose filler particles are often chemically inert to the environment around them. Because of this, filler particles can often only be formed/shaped together with a binder. Suitable binders are, for example, organic and inorganic binders. One exception to this are fillers which are capable of setting to form solid materials by reacting with a reactant present in the surrounding environment. Examples of such fillers are gypsum in the form of anhydrite and various lime compounds which are capable of curing by reacting with water or carbon dioxide from the surrounding air.

Whereas the self-setting fillers mentioned above generally cure to form brittle hard materials, a distinctly broader spectrum of physical and

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chemical properties can often be achieved by replacing the inorganic materials with polymers. However, disadvantages of using polymers only generally include increased production costs by comparison with inorganic materials and their lower hardness and chemical resistance, particularly in  
5 regard to fire resistance and flameproofing.

In this connection, the building supplies industry in particular has a steadily increasing demand for new materials which combine the positive properties of fillers, such as their chemical resistance, their resistance to heat, their availability in large quantities and their low price, with those of  
10 the polymers. The demand ranges from surface coating compositions or adhesives, which are generally applied as a thin layer to surfaces of various kinds, through surfacing and sealing compounds to plastics of the type used, for example, as a sheathing for power cables or as water conduits.

For example, Wirsching, Hüller, Hofmann and Pürzer describe the use of fillers of WDP gypsum in ZKG INTERNATIONAL, No. 5, 1995 (Vol. 48), pp. 241-256 (Bauverlag GmbH). The article in question describes in particular the use of WDP gypsum from coal-fired power stations in adhesives, paints and plastics. Before being used as a filler, the gypsum is  
15 finely ground so that it has a mean particle diameter of about 8 to 12  $\mu\text{m}$  and the upper cut of the particle diameter is at about 25 to 50  $\mu\text{m}$ .  
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JP 76-139114 relates to the use of WDP gypsum as a pigment in coating compositions. This document describes a composition of titanium dioxide, WDP gypsum, aluminium silicate, ethylene/vinyl acetate  
25 copolymer, polyvinyl acetate, thickener and water as a white emulsion which is suitable as a coating composition.

Filler-containing polymer materials are often marketed and processed in the form of aqueous dispersions. Unfortunately, such materials frequently have serious disadvantages during and after  
30 processing. Firstly, the viscosity of the dispersions often cannot be

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adjusted to a value suitable for processing. Secondly, after processing and generally after drying, the filler-containing polymer material applied undergoes a distinct change in volume in relation to its volume at the time of application. This change in volume can often correspond to the extent to  
5 be expected as a result of evaporation of the water present in the dispersion.

Behavior such as this (often referred to as "shrinkage" or "contraction") is undesirable, above all for filler-containing polymer dispersions which are supposed to perform a "filling" function. Thus, in the  
10 case of surface coatings for example, importance is often attributed to the levelling of any structural unevenness of the substrate. Surfacing or sealing compounds, for example, are expected to fill the void to be sealed or filled - even after drying of the compound introduced - to the full extent to which the dispersion was originally introduced.

15 In addition, the contraction of surfacing compounds frequently leads during drying of the compound to the formation of cracks therein which, besides the generally noticeable visual disadvantages, often represent starting points for the penetration of corrosive compounds or moisture. The visual impression and the stability of the region thus filled are often  
20 drastically impaired as a result.

Filler-containing polymer materials are often used as adhesives, more particularly emulsion-based adhesives. Unfortunately, many adhesives of this type lack elasticity which is often a disadvantage to the stability of the adhesive bond under stress.

*SUMMARY OF THE INVENTION*  
25 Now, the problem addressed by the invention was to remedy these disadvantages. This problem has been solved by a polymer composition which, besides a water-soluble or water-dispersible polymer or a mixture of two or more such polymers, contains filler particles in the form of gypsum particles from waste-gas desulfurization plants of which the particle size  
30 distribution has a mean diameter ( $\times 50$ ) of about 13 to about 500  $\mu\text{m}$ .

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*DETAILED DESCRIPTION OF THE INVENTION*

Accordingly, the present invention relates to a preparation containing a water-soluble polymer or a water-dispersible polymer or a mixture of two or more thereof and filler particles, the filler particles being gypsum particles from waste-gas desulfurization plants (WDP gypsum) with a value  
5 for the particle size distribution x50 of 13 to 500  $\mu\text{m}$  (as measured with a Sympatec Helos H0720 in isopropanol).

In the context of the invention, a "preparation" is any mixture which contains the constituents mentioned above. It may be a mixture which is already in a form suited to the application envisaged (for example already  
10 provided with a suitable quantity of water) or which first has to be brought by the user into a form suitable for application, for example a powder dispersible in water.

A "water-soluble polymer or water-dispersible polymer" in the context of the present invention is a polymer which, in the form of a mixture  
15 with water, forms a molecularly disperse solution, a substantially stable gel or colloid or a substantially stable dispersion. In the context of the present invention, it does not matter whether the stability of the aqueous preparations mentioned derives from the properties of the polymer itself or is supported by such additives as emulsifiers, stabilizers, gel formers or the  
20 like.

The preparation according to the invention may contain, for example, only a certain water-soluble polymer or a certain water-dispersible polymer. However, the preparation may equally well contain a mixture of two or more water-soluble polymers or a mixture of two or more  
25 water-dispersible polymers. According to the invention, the preparation may also be a mixture of one or more water-soluble polymers and one or more water-dispersible polymers.

The water solubility or self-dispersibility of polymers can be based, for example, on the presence of anionic or cationic groups such as are  
30 normally present in polymers to achieve such an effect. Solubility or

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dispersibility in water can also be obtained by arranging nonionic groups on a polymer suitable for use in accordance with the invention, these nonionic groups leading to water solubility or water dispersibility of the polymer.

Suitable cationic groups are, for example, quaternized amino groups; suitable anionic groups are, in particular, acid groups.

For example, water-soluble polymers obtainable by polymerization of monomer constituents which impart water solubility to the polymers may be used for the purposes of the present invention. Such polymers include, for example, the polymers of acrylic acid and the polymers obtainable by polyaddition of alkylene oxides. Polymers which are "self-dispersible" in water are also suitable. "Self-dispersible polymers" are understood to be polymers which form a substantially stable dispersion in water without the addition of emulsifiers. In general, polymers such as these contain, for example, carboxylic acid groups, sulfonic acid groups, phosphonic acid groups or chain segments of polyethylene oxide or a mixture of two or more of the functional groups mentioned as functional groups.

Polymers which are neither soluble in water nor "self-dispersible" may be converted into a substantially stable emulsion or dispersion in water, for example using commercially available emulsifiers or dispersants.

Suitable polymers are, for example, polyurethanes, polyacrylates, polymethacrylates, polyvinyl esters, polystyrene and sulfonated polystyrene, polybutadiene and sulfonated polybutadiene, polyamides, polyesters and polyvinyl chloride. Other suitable polymers are corresponding co- and ter-polymers, such as ethylene/vinyl acetate copolymers (EVA), styrene/butadiene copolymers (SBR), styrene/acrylonitrile polymers (SAN), styrene/acrylate copolymers and the like. Another preferred embodiment is characterized by the use of, for example, polymers obtainable from the polymerization of acrylates or from the co- and ter-polymerization of acrylates with acrylonitrile, vinyl esters, maleates, acrylic acid, styrene and the like. Polymers such as these and

polymers dispersions resulting from them are comprehensively described, for example, in "Encyclopaedia of Polymer Science and Technology" (Editors: Mark, Bikales, Overberger, Menges, 2nd Edition, 1989, Wiley, New York, 17, pp. 406-409).

5           In another preferred embodiment, water-soluble or water-dispersible polymers or mixtures thereof already present in dissolved or dispersed form are used for the production of the preparation according to the invention. These polymers are in particular aqueous dispersions of synthetic polymers of the type mentioned above, more particularly polyurethanes,  
10 poly(meth)acrylates, polyvinyl esters, polystyrene, polybutadiene, polyamides or polyvinyl chloride, or mixtures of two or more thereof. The corresponding co- and ter-polymers, styrene/butadiene, styrene/acrylates may also be used for the purposes of the present invention, as may natural latices. The dispersions usable in accordance with the invention may be  
15 prepared, for example, by suspension or emulsion polymerization of the corresponding monomers. Secondary dispersions - obtainable by dispersing a polymer melt in a suitable medium - may also be used.

Polymer dispersions which are generally obtainable commercially in large quantities as the outcome of suspension or emulsion polymerizations and which are used, for example, as binders for emulsion paints or  
20 emulsion-based adhesives (see, for example, Römpp Chemie-Lexikon, Vol. 2, Thieme-Verlag, 1990, pp. 1010-1011 and the literature cited therein) are preferably used. Unsaturated, radical-polymerizable compounds, such as acrylates and methacrylates, dienes or olefins, or mixtures of two or  
25 more thereof are particularly suitable as monomers for such polymer dispersions. The suspension or emulsion polymerization is described, for example, in "Ullmann's Enzyklopädie der technischen Chemie" (Vol. A21, 5th Edition, VCH, 1987), reference being expressly made to this literature reference.

30           Polymer dispersions used in a preferred embodiment of the present

invention are based on polyvinyl esters, such as polyvinyl acetate, and co- and ter-polymers of the vinyl esters with monomers, such as ethylene, acrylates or methacrylates or maleic acid mono- and diesters, or mixtures of two or more thereof. Monomers which lead to dispersions suitable for use in accordance with the present invention are described, for example, in Ullmann's Enzyklopädie der technischen Chemie (Vol. A22, 1993, VCH, pp. 1-15). Dispersions prepared on the basis of such monomers are described, for example, in "Handbook of Additives" (3rd Edition, Chapman and Hall, pp. 381-399). Reference is hereby expressly made to the cited literature references.

Polymers suitable in a preferred embodiment are, for example, the copolymers of vinyl acetate, vinyl propionate or VeoVa® 9 or 10 with other comonomers. VeoVa® 9 and 10 are the vinyl esters of tert. carboxylic acids (Versatic® Acid 9 or 10) for copolymerization with, for example, vinyl acetate for emulsion paints, plasters, concrete additives, paper and textile coatings, emulsion-based adhesives and paints (manufacturer: Deutsche Shell Chemie). Particularly suitable polymers are co- and ter-polymers containing vinyl acetate/dibutyl maleate, vinyl acetate/n-butyl acrylate, vinyl acetate/2-ethylhexyl acrylate, vinyl acetate/n-butyl acrylate/N-hydroxymethyl acrylamide, vinyl acetate/crotonic acid, vinyl acetate/VeoVa® 10, vinyl acetate/VeoVa® 10/acrylic acid, vinyl acetate/VeoVa® 10/n-butyl acrylate, vinyl acetate/N-hydroxymethyl acrylamide, vinyl acetate/vinyl laurate, vinyl acetate/vinyl laurate/vinyl chloride, vinyl acetate/ethylene/vinyl chloride, vinyl acetate/ethylene/acrylate, vinyl acetate/ethylene/acrylamide, vinyl acetate/ethylene/N-hydroxymethyl acrylamide, vinyl propionate, vinyl propionate/vinyl chloride, vinyl propionate/tert.butyl acrylate, VeoVa® 10/vinyl chloride, VeoVa® 10/styrene/acrylate, VeoVa® 10/styrene/maleate, VeoVa® 10/styrene/acrylate/maleate and VeoVa® 10/VeoVa® 9/methyl methacrylate/butyl acrylate, VeoVa® 10 being completely or partly

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replaceable by VeoVa® 9.

Another preferred embodiment of the invention is characterized by the use of polymer dispersions based on poly(meth)acrylates and co- and ter-polymers of the (meth)acrylates with such monomers as acrylonitrile, vinyl esters, maleates, acrylic acid and styrene. Corresponding polymer dispersions are comprehensively described, for example, in "Emulsion Polymerisation and Emulsion Polymers" (1997), John Wiley, pp. 619-655, New York) and are regarded as part of the present disclosure.

Another preferred embodiment of the invention is characterized by the use of commercially obtainable dispersions, such as DL 345 (manufacturer: Dow Latex) or Acronal® DS 3518 (manufacturer: BASF AG).

Aqueous polymer dispersions based on conjugated dienes, such as chloroprene or butadiene, and copolymers of these dienes with unsaturated compounds, such as styrene or acrylonitrile, may also be used in accordance with the present invention. Such dispersions are known and are described, for example, in "Emulsion Polymerisation and Emulsion Polymers" (1997), John Wiley, pp. 521-561, New York). Reference is hereby specifically made to the cited literature reference.

Besides the polymers mentioned, monomers with additional functional groups, such as N-methylol acrylamide, hydroxypropyl acrylate, (meth)acrylic acid or a mixture of two or more of the compounds mentioned, may be used for preparing the polymer dispersions usable in accordance with the invention.

The preparations according to the invention contain at least WDP gypsum particles with a value for the particle size distribution x50 of 13 to 500 µm (as measured with a Sympatec Helos H0720) as filler.

WDP gypsum particles differing in their dimensions are formed in different waste-gas desulfurization plants in dependence upon the technical parameters. It has been found in accordance with the present invention



that the advantages according to the invention can be obtained with WDP gypsum particles having the above-mentioned values for the particle size distribution.

In general, various processes may be used to measure a particle size distribution and the corresponding value x50. Typical processes include, for example, the sieve process where a certain quantity of particles is sieved through sieves with different mesh widths. The total quantity of particles is thus divided into fractions with different particle diameters of which the quantity is expressed as a percentage of the total weight of the particles investigated. Other methods for determining particle size distribution are, for example, light scattering and Fraunhofer diffraction. In the context of the present invention, the particle size distribution of the WDP gypsum particles was determined by the Fraunhofer diffraction technique using a Sympatec Helos H0720. The particle size distribution was measured on a suspension in isopropanol. The following particle size distribution data are based on measurements by the technique mentioned but are not confined to such measurements. The advantages according to the invention can generally be obtained with any WDP gypsum particles whose particle size distribution is largely within the range mentioned, irrespective of the measuring system.

In a preferred embodiment of the present invention, the WDP gypsum particles have a lower limit to the particle size x50 of at least about 25  $\mu\text{m}$ . In another preferred embodiment, the value for the particle size distribution x50 is about 30 to 250  $\mu\text{m}$ . Good results can be obtained, for example, where the values for the particle size distribution x50 are in the range from about 35 to about 200 or of the order of 150  $\mu\text{m}$ . Another preferred embodiment of the invention is characterized by the use of WDP gypsum particles with a x50 value for the particle size distribution of from about 40 to about 120  $\mu\text{m}$ , for example in the range from about 60 to about 110  $\mu\text{m}$  and more particularly in the range from about 80 to about 100  $\mu\text{m}$ .

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It has also been found to be of advantage for the filler particles to have a granular or rodlet-like form.

The WDP gypsum particles used as filler particles in accordance with the invention show their advantages according to the invention even  
5 when they are used as sole filler. In this case, WDP gypsum particles with a x50 value for the particle size distribution in the range from about 13 to about 110  $\mu\text{m}$  and more especially in the range from about 35 to about 80  $\mu\text{m}$  are used in a preferred embodiment of the invention.

In another preferred embodiment of the present invention, the WDP  
10 gypsum particles are used in admixture with at least one other type of inorganic filler particles.

In the context of the invention, the expression "one other type" of inorganic filler particles is intended to encompass any filler particles which differ from WDP gypsum particles either in their chemical composition, in  
15 their predominant spatial form (for example their crystal form) or in a combination of two or more of the features mentioned. In a preferred embodiment of the present invention, filler particles which differ at least in the x50 value of their particle size distribution from the corresponding value of the particle size distribution of the WDP gypsum particles are used as  
20 the other type of inorganic filler particles.

Suitable other types of inorganic filler particles are, for example, any inorganic filler particles inert to the other substances present in the preparation according to the invention. So far as the other type of inorganic filler particles is concerned, there are no restrictions on the x50  
25 value for the particle size distribution. For example, filler particles with a x50 value for the particle size distribution in the range from about 0.01 to about 500  $\mu\text{m}$  may be used in accordance with the present invention.

Suitable other types of inorganic filler particles are, for example, filler particles of andalusite, sillimanite, kyanite, pyrophyllite, imigolite or  
30 allophane. Also suitable are compounds based on sodium aluminates or

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calcium silicates and minerals, such as silica, calcium sulfate (gypsum) which does not come from waste-gas desulfurization plants in the form of anhydrite, semihydrate or dihydrate, silica flour, silica gel, barium sulfate, titanium dioxide, zeolites, leucite, potash feldspar, biotite, the group of  
5 soro-, cyclo-, ino-, phyllo- and tectosilicates, the group of poorly soluble sulfates, such as gypsum, anhydrite or heavy spar, and calcium minerals, such as calcite or chalk ( $\text{CaCO}_3$ ). According to the invention, the inorganic materials mentioned may be used individually as another type of inorganic filler particles. However, a mixture of two or more of the compounds  
10 mentioned may equally well be used. A preferred embodiment of the invention is characterized by the use of calcite, kaolin, dolomite, silica flour and gypsum ( $\text{CaSO}_4 \cdot 2 \text{H}_2\text{O}$ ).

In another preferred embodiment of the present invention, the filler particles of the other type have a  $x_{50}$  value for the mean particle size  
15 distribution in the range from about 1 to about 120  $\mu\text{m}$ , for example in the range from about 3 to about 60  $\mu\text{m}$  or in the range from about 60 to about 90  $\mu\text{m}$ .

Organic filler particles which cannot readily be assigned to the water-soluble or water-dispersible polymers are also suitable for use as  
20 another type of filler particles. These include in particular the finely ground plastic powders obtained in the recycling of plastics, more particularly the plastic powders obtainable from the fine grinding of highly crosslinked elastomeric or thermoset polymers. One example of such powders is the rubber powder obtained, for example, in the fine grinding of car tires.

25 Where the preparation according to the invention contains some filler particles of the other type either individually or in the form of a mixture of two or more thereof, the ratio by weight of WDP gypsum particles to filler particles of the other type is about 1:1,000 to about 1,000:1. Good results can be obtained, for example, if the ratio by weight of WDP gypsum  
30 particles to filler particles of the other type is about 1:10 to about 10:1 and

more particularly about 5:1 to about 1:5.

In another preferred embodiment of the present invention, the filler component of the preparation contains at least about 0.5% by weight and, more particularly, at least about 1% by weight of WDP gypsum particles. In

5 another preferred embodiment of the present invention, the percentage content of WDP gypsum particles is at least about 10% by weight or at least about 20% by weight. Good results are also obtained with percentage contents of WDP gypsum particles of at least about 30, 40 or 50% by weight or more, for example about 60 to 80% by weight or at least  
10 about 90% by weight.

In a preferred embodiment, the preparation according to the invention contains at least about 20% by weight of filler particles (WDP gypsum particles or other filler particles or a mixture thereof). A larger percentage of filler particles, for example about 30, 40 or 50 to 99% by  
15 weight, for example about 60 to 90% by weight, may also be added.

The compositions according to the invention may be present as a ready-to-use aqueous dispersion, i.e. they may contain the water-soluble or water-dispersible polymer or the mixture of two or more such polymers and the fillers together with water. According to the invention, however, the  
20 compositions may equally well contain no water or only a little water, i.e. for example may be present as a dry powder or as a paste of low water content. According to the invention, the preparation according to the invention may also be present as a water-free paste, a non-aqueous liquid, for example a solvent or another constituent of the preparation according to  
25 the invention, being used to establish the paste-like properties. Corresponding powders or pastes are of advantage, for example, when the actual preparation of an aqueous dispersion intended for use is to be left to the user himself/herself. In a preferred embodiment of the present invention, the preparation according to the invention is present in the form  
30 of a powder (redispersion powder).

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If WDP gypsum particles are to be used as filler in accordance with the present invention, it is important to test the stability of the polymer dispersion used to calcium ions. In such a case, stability may have to be restored or improved by adding another emulsifier or dispersant or a mixture of two or more emulsifiers or dispersants.

Besides the polymer(s) and the filler particles, the preparation according to the invention may contain other ingredients. If the preparation according to the invention is intended to be ready-to-use, it may contain water. Depending on the application envisaged for the preparation according to the invention, the water content may vary between about 0 and 49% by weight.

Basically, water contents (based on the preparation as a whole) of about 0 to 49% by weight are suitable.

In addition to the constituents mentioned, the composition according to the invention may also contain one or more other additives.

Suitable other additives are, for example, emulsifiers, dispersants, stabilizers, defoamers, antioxidants, photostabilizers, pigment dispersants and the like.

The present invention also relates to a process for the production of a preparation containing a water-soluble polymer or a water-dispersible polymer or a mixture thereof and WDP gypsum particles with a particle size x50 of 13 to 500  $\mu\text{m}$  (as measured with a Sympatec Helos H0720 in isopropanol) or a mixture of WDP gypsum particles and at least one other type of inorganic filler particles, characterized in that at least one water-soluble polymer or at least one water-dispersible polymer or a mixture of two or more thereof or an aqueous dispersion containing one or more such polymers is mixed with WDP gypsum particles having a particle size x50 of 13 to 500  $\mu\text{m}$  (as measured with a Sympatec Helos H0720 in isopropanol) or a mixture of WDP gypsum particles and at least one other type of inorganic filler particles and optionally with water and one or more other

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additives in one or more mixing steps in any order and with any time intervals between individual mixing steps.

The invention is illustrated by the accompanying drawings wherein:

Figure 1 is an electron micrograph of WDP gypsum with a granular  
5 to rodlet-like particle form.

Figure 2 shows the size distribution of the filler particles of a WDP gypsum used by way of example as filler.

The present invention also relates to the use of WDP gypsum particles having a x50 value of the particle size distribution of 13 to 500  $\mu\text{m}$   
10 (as measured with a Sympatec Helos H0720 in isopropanol) for the production of surface coatings, surfacing compounds, sealing compounds, adhesives or moldings with a content of water-soluble or water-dispersible polymers.

In a preferred embodiment of the invention, the WDP gypsum particles have a x50 value for the particle size distribution of 30 to 250  $\mu\text{m}$ .  
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The present invention also relates to the use of a mixture of inorganic filler particles containing WDP gypsum particles with a x50 value for the particle size distribution of 30 to 250  $\mu\text{m}$  and at least one other type of inorganic filler particles as a filler in polymer dispersions.

20 The invention is illustrated by the following Examples.

### Examples

#### Example 1: elasticity of emulsion-based adhesives

25 Compositions according to the invention show favorable elongation behavior when used as an emulsion-based adhesive.

Formulation:

acrylate dispersion (for example Acronal® DS 3518, BASF)	55 g
pigment dispersant (for example Pigmentverteiler A, BASF)	2 g

30 Fillers:

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- A: fine calcite filler (for example Omyacarb 5GU, D50 value 6  $\mu\text{m}$ ; Omya)
- B: unground WDP gypsum (for example from Rethmann, D50 value 40  $\mu\text{m}$ )
- C: ground natural gypsum (for example Alabaster Brilliantweiß, D50 value 11  $\mu\text{m}$ ; Börgardts)
- D: coarse calcite filler (for example 50:50 mixture of Omyacarb 130 AL/Omyacarb 40 GU, D50 value (mixture) 88  $\mu\text{m}$ ; Omya)

Same-weight Filler Exchange							
Filler mixture					Max. force [kN/mm <sup>2</sup> ]	Elongation at max. force [%]	Elongation at break [%]
A [%]	A [g]	B [g]	C [g]	D [g]			
0	0	84	-	-	0.01	260	2840
50	42	42	-	-	0.02	680	1060
100	84	0	-	-	0.03	190	830
0.02	0	-	84	-	0.02	90	1050
0.03	42	-	42	-	0.03	140	900
100	84	-	0	-	0.03	190	830
0	0	-	-	125	0.01	420	2330
20	25	-	-	100	0.02	390	1500
50	62.5	-	-	62.5	0.02	340	1250
80	100	-	-	25	0.03	280	780
100	125	-	-	0	0.03	130	630

Example 2: volume shrinkage of surfacing compounds as a function of the filler composition

Visual evaluation

Formulation:

Styrene/acrylate dispersion (for example DL 345 from Dow Latex) 110 g

Pigment dispersant (for example Pigmentverteiler A, BASF) 4 g

Filler:

Filler used in 50 or 100% same-volume or same-weight exchange for filler

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A (visual evaluation: -)

- A: fine calcite filler (for example Omyacarb 5GU, D50 value 6  $\mu\text{m}$ ; Omya)
- B: unground WDP gypsum (for example from Rethmann, D50 value 40  $\mu\text{m}$ )
- C: ground natural gypsum (for example Alabaster Brilliantweiß, D50 value 11  $\mu\text{m}$ ; Börgardts)
- D: coarse calcite filler (for example 50:50 mixture of Omyacarb 130 AL/Omyacarb 40 GU, D50 value (mixture) 88  $\mu\text{m}$ ; Omya)
- E: unground WDP gypsum (for example No. 1 from ProMineral, D50 value 36  $\mu\text{m}$ )
- F: unground WDP gypsum (for example No. 2 from ProMineral, D50 value 96  $\mu\text{m}$ )
- G: medium-coarse calcite filler (for example Omyacarb 40 GU, D50 value 44  $\mu\text{m}$ ; Omya)

+: no visible volume shrinkage, no cracks

±: slight volume shrinkage/crack formation

-: distinct volume shrinkage/crack formation

Filler =	Same-weight Filler Exchange		Same-volume Filler Exchange	
	50% by weight X	100% by weight X	50% by vol. X	100% by vol. X
B	±	+	-	±
C	±	±	-	-
D	-	-	-	-
E	-	+	-	+
F	+	+	+	+
G	-	-	-	-

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Example 3: tensile shear strength (wood-to-wood bonds) and viscosity behavior of emulsion-based adhesives

Formulation:

Styrene/acrylate dispersion (for example DL 345 from Dow Latex) 110 g

Pigment dispersant (for example Pigmentverteiler A, BASF) 4 g

Fillers:

A: fine calcite filler (for example Omyacarb 5GU, D50 value 6  $\mu\text{m}$ ; Omya)

B: unground WDP gypsum (for example from Rethmann, D50 value 40  $\mu\text{m}$ )

C: ground natural gypsum (for example Alabaster Brilliantweiß, D50 value 11  $\mu\text{m}$ ; Börgardts)

D: coarse calcite filler (for example 50:50 mixture of Omyacarb 130 AL/Omyacarb 40 GU, D50 value (mixture) 88  $\mu\text{m}$ ; Omya)

Same-weight Filler Exchange						
Filler Mixture					Tensile shear strength [N/mm <sup>2</sup> ]	Viscosity [scale units]
A [%]	A [g]	B [g]	C [g]	D [g]		
0	0	250	-	-	2.1	100
50	125	125	-	-	4.7	38
100	250	0	-	-	2.7	210
0	0	-	250	-	3.7	100
50	125	-	125	-	3.0	94
100	250	-	0	-	2.7	210
0	0	-	-	250	3.1	36
20	50	-	-	200	3.2	41
50	125	-	-	125	3.8	39
80	200	-	-	50	2.9	135
100	250	-	-	0	2.7	210

Example 4: Tensile shear strength (wood-to-wood bonds) and viscosity behavior of emulsion-based adhesives containing WDP gypsum with different particle size distributions

## Formulation:

Styrene/acrylate dispersion (for example DL 345 from Dow Latex) 110 g

Pigment dispersant (for example Pigmentverteiler A, BASF) 4 g

## Fillers:

A: fine calcite filler (for example Omyacarb 5GU, D50 value 6 µm;  
Omya)E: unground WDP gypsum (for example No. 1 from ProMineral, D50  
value 36 µm)F: unground WDP gypsum (for example No. 2 from ProMineral, D50  
value 96 µm)G: medium-coarse calcite filler (for example Omyacarb 40 GU, D50  
value 44 µm; Omya)

Same-weight Filler Exchange						
Filler Mixture					Tensile shear strength [N/mm <sup>2</sup> ]	Viscosity [scale units]
A [%]	A [g]	E [g]	F [g]	G [g]		
0	0	250	-	-	2.2	90
20	50	200	-	-	2.9	37
50	125	125	-	-	3.5	27
80	200	50	-	-	3.4	43
100	250	0	-	-	2.7	210
205	0	-	250	-	1.1	120
164	50	-	200	-	1.6	60
102	125	-	125	-	2.7	19
41	200	-	50	-	4.3	42
0	250	-	0	-	2.7	210
0	0	-	-	250	3.4	38
20	50	-	-	200	3.5	45
50	125	-	-	125	3.1	56
80	200	-	-	50	2.5	80
100	250	-	-	0	2.7	210